Influence of the $L2_1$ ordering degree on the magnetic properties in Co_2MnSi Heusler films

O. Gaier,[1] J. Hamrle,[1] S. J. Hermsdoerfer,[1]

B. Hillebrands,[1] Y. Sakuraba,[2] M. Oogane,[2] and Y. Ando[2]

[1]*Fachbereich Physik and Forschungsschwerpunkt MINAS,*

Technische Universität Kaiserslautern,

Erwin-Schrödinger-Straße 56, D-67663 Kaiserslautern, Germany

[2]*Department of Applied Physics, Graduate School of Engineering,*

Tohoku University, Aoba-yama 6-6-05,

Aramaki, Aoba-ku, Sendai 980-8579, Japan

(Dated: February 1, 2008)

Abstract

We report on the influence of the improved $L2_1$ ordering degree on the magnetic properties of Co_2MnSi Heusler films. Different fractions of the $L2_1$ phase are obtained by different post-growth annealing temperatures ranging from 350 °C to 500 °C. Room temperature magneto-optical Kerr effect measurements reveal an increase of the coercivity at an intermediate annealing temperature of 425 °C, which is a fingerprint of an increased number of pinning centers at this temperature. Furthermore, Brillouin light scattering studies show that the improvement of the $L2_1$ order in the Co_2MnSi films is correlated with a decrease of the saturation magnetization by about 9 %. The exchange stiffness constant of Co_2MnSi, however, increases by about 8 % when the $L2_1$ order is improved. Moreover, we observe a drop of the cubic anisotropy constant K_1 by a factor of 10 for an increasing amount of the $L2_1$ phase.

I. INTRODUCTION

The performance of spin dependent devices like magnetic tunnel junctions (MTJs) and spin valves can be considerably improved if the conduction electrons are spin polarized. Half-metallic ferromagnets (HMFs) are promising candidates for these applications as they are predicted to exhibit 100 % spin polarization at the Fermi level[1,2]. Among different HMFs the Heusler compound Co_2MnSi has attracted particular interest due to its high Curie temperature T_C of 985 K and a large energy gap of 0.4 eV in the minority spin band[3,4].

Recently, the fabrication and the characterization of MTJs with one electrode consisting of Co_2MnSi has been reported[5,6,7,8]. For example, in a Co_2MnSi/Al-O/$Co_{75}Fe_{25}$ structure Sakuraba *et al.* obtained a tunneling magnetoresistance (TMR) ratio of 159 % at 2 K and 70 % at room temperature (RT)[6]. According to Julliere's model, the spin polarization at the Co_2MnSi interface is 89 % at 2 K[7,8]. This is comparable to a spin polarization of 61 % previously reported by Schmalhorst *et al.* at 10 K. More recently, an increase of the TMR ratio to an extremely high value of 570 % at 2 K was achieved in a Co_2MnSi/Al-O/Co_2MnSi structure by integrating a second Co_2MnSi electrode[7]. At RT however, no further enhancement of the TMR ratio could be observed. This ratio was found to be 67%[7] and is therefore similar to the value obtained in the aforementioned Co_2MnSi/Al-O/$Co_{75}Fe_{25}$ structure[6].

In contrast to the theoretically expected behavior the highest value of spin polarization in Co_2MnSi films at RT reported so far is only 12 %. This value was measured by means of spin-resolved photoemission spectroscopy on films epitaxially grown onto a GaAs(001) substrate[9]. The discrepancy between the experiment and the expected half-metallic behaviour is mainly attributed to structural defects or site-disordering in the Co_2MnSi lattice. For example, according to *ab initio* calculations performed by Picozzi *et al.*[10], Co-antisite disorder leads to defect-induced states in the gap of the minority spin band which in turn leads to a reduction of the spin polarization.

In this work, we investigate how the magnetic properties of thin Co_2MnSi films are modified when the crystal structure is gradually improved. For this purpose, room temperature magneto-optical Kerr effect and Brillouin light scattering studies were performed on Co_2MnSi thin films with increasing fractions of the $L2_1$ phase. Here, $L2_1$ denotes a perfectly ordered crystal structure consisting of four interpenetrating fcc lattices, each of which is occupied by a different kind of atoms. In contrast, the crystal structure is referred to as

B2 when Mn and Si atoms are randomly disordered and as A2 when a random disorder between all elements is existent. In the following, we describe the preparation and the structural characterization of the Co_2MnSi samples before presenting the results of our magnetic investigations.

II. SAMPLE PREPARATION AND STRUCTURAL CHARACTERIZATION

The investigated Co_2MnSi films of 30 nm thickness and (100) orientation were epitaxially grown by inductively coupled plasma-assisted magnetron sputtering onto a MgO(100) substrate covered by a 40 nm thick Cr(100) buffer layer. All Co_2MnSi films were capped by a 1.3 nm Al protection layer. After the deposition, the Co_2MnSi films were annealed at different annealing temperatures T_a ranging from 350 °C to 500 °C to provide samples with different degrees of the $L2_1$ order.

The analysis of the crystal structure was carried out by means of X-ray diffraction (XRD) using Cu-K_α radiation. The X-ray θ-2θ-scans obtained from the Co_2MnSi samples annealed at different temperatures T_a are summarized in Fig. 1. For all studied films clear peaks corresponding to the (200) and (400) reflections of Co_2MnSi are observed in the specular geometry indicating perfect epitaxial (100) growth. Furthermore, pole figure scans of all Co_2MnSi samples show the presence of the (111) peak that is characteristic for the $L2_1$ structure. An example of such a pole figure scan is presented in Fig. 2(a) for the Co_2MnSi film annealed at 450 °C. Figure 2(b) shows the pole figure of the corresponding (220) fundamental reflections. The total integrated intensities of the (111) and (220) reflections obtained from those pole figure scans were used to estimate the degree of the $L2_1$ ordering in the Co_2MnSi films annealed at different T_a. For this purpose, the long-range order parameter S_{L2_1} was calculated using the following expression:

$$S_{L2_1}^2 = \frac{I_{obs}(111)/I_{obs}(220)}{I_{cal}(111)/I_{cal}(220)}. \tag{1}$$

Here, I_{obs} and I_{cal} denote the peak intensities obtained from the experiment and powder pattern simulations, respectively. Correction factors such as the multiplicity and the film thickness factors[11] were taken into account when evaluating I_{cal} from the simulated intensities.

The dependence between the long-range order parameter S_{L2_1} and the annealing tem-

perature T_a is presented in Fig. 3. The values of S_{L2_1} increase from approximately 0.6 to about 0.9 with increasing T_a except for the sample annealed at $T_a = 475\,^{\circ}$C. This feature is probably related to a statistical fluctuation of the experimental parameters during the preparation procedure. For this reason, the Co_2MnSi film with $T_a = 475\,^{\circ}$C will not be considered further in the analysis of the experimental data. $S_{L2_1} = 1$ in the case of a perfect order on the Mn and Si sites. Therefore, we estimate that the Co_2MnSi film annealed at $T_a = 500\,^{\circ}$C is 80-90 % L2_1 ordered. The reduction of S_{L2_1} with decreasing T_a corresponds to the decreasing fraction of the L2_1 phase inside the film.

III. MAGNETO-OPTICAL KERR EFFECT STUDIES

The influence of the L2_1 ordering degree in Co_2MnSi films on their coercivity was studied by means of a standard magneto-optical Kerr effect (MOKE) set-up in longitudinal geometry, i.e., the external magnetic field was applied in-plane and parallel to the plane of incidence. The sample was investigated at room temperature by s-polarized laser light with a wavelength of $\lambda = 670$ nm and an angle of incidence of 45°. MOKE hysteresis loops were measured at different sample orientations α varied from 0° to 360° in steps of 1° with α being the angle between the in-plane [110] direction of the Co_2MnSi film and the plane of incidence.

The hysteresis loops of all investigated Co_2MnSi films acquired from MOKE measurements reveal a strongly asymmetric shape depending on the sample orientation with respect to the direction of the applied magnetic field. For example, Fig. 4 shows the hysteresis curves of the Co_2MnSi film annealed at $T_a = 425\,^{\circ}$C obtained for the different sample orientations $\alpha = 0°$, 42° and 45°. The observed asymmetry results from the quadratic MOKE (QMOKE) contribution which is proportional to quadratic terms in magnetization to the detected signal, as already reported for various other materials[12,13,14,15]. By symmetrization and antisymmetrization of experimental loops (i.e., by subtracting or adding the loop branches for increasing and decreasing H values) it is possible to separate linear, i.e., longitudinal MOKE (LMOKE) and QMOKE contributions[14], as also shown in Fig. 4.

The LMOKE curves are used to determine the values of the coercive field at different sample orientations and for different annealing temperatures. The resulting polar plots (Fig. 5) reveal a four-fold magnetic anisotropy for all investigated samples, reflecting the

cubic symmetry of the crystal structure. Moreover, in all cases sharp peaks in H_C appear when the applied magnetic field is aligned parallel to a hard direction. These peaks originate from a checkerboard domain pattern which occurs during hard axis magnetization reversal processes as reported for 80 nm thick $Co_2Cr_{0.6}Fe_{0.4}Al$ Heusler films[16]. The variation of the coercivity with T_a is shown in Fig. 6 for three different sample orientations. A maximum of H_C is clearly visible for the Co_2MnSi film annealed at 425 °C independently from the sample orientation α. The observed increase of H_C is probably related to an enhanced number of pinning centers. In the Co_2MnSi samples investigated here, such an increased number of pinning centers might originate from the diffusion of Cr atoms from the buffer layer into the Co_2MnSi film.

IV. BRILLOUIN LIGHT SCATTERING STUDIES

The Brillouin light scattering (BLS) technique was used to study changes in the spin wave frequencies of Co_2MnSi films related to an increasing amount of the $L2_1$ phase. All BLS spectra were measured at room temperature at a transferred wave vector of $q_\parallel = 1.67\,\mathrm{cm}^{-1}$ using laser light with a wavelength of $\lambda = 532\,\mathrm{nm}$. The magnetization vector was oriented parallel to an in-plane easy axis direction of the Co_2MnSi samples and perpendicular to the wave vector of the incident light ($\boldsymbol{M}\perp\boldsymbol{k}$). Details of the BLS spectrometer as well as a description of the data acquisition have been published elsewhere[17].

For the geometry of the BLS set-up used here ($\boldsymbol{M}\perp\boldsymbol{k}$) two kinds of spin waves can be excited: dipole dominated magnetostatic surface waves, also called Damon-Eshbach modes (DE), and exchange dominated perpendicular standing spin waves (PSSW). DE modes are characterized by an exponential decay of the amplitude of the dynamic magnetization over the film thickness d and their nonreciprocal behavior (i.e., propagation is possible for either positive or negative direction of the wave vector, but not for both). In the case of week exchange contribution and negligible anisotropies, the wave vector dependence of DE mode frequency is given by[18]

$$\nu_{DE}(q_\parallel) = \frac{\gamma}{2\pi}\left[H(H + 4\pi M_S) + (2\pi M_S)^2(1 - e^{-2q_\parallel d})\right]^{\frac{1}{2}}, \qquad (2)$$

where γ is the modulus of the gyromagnetic ratio for the electron spin, H the externally applied magnetic field, $4\pi M_S$ the saturation magnetization and $q_\parallel$ the in-plane wave vector

of the spin wave. PSSW modes are a superposition of two waves propagating in opposite directions with a wave vector $q \approx p\pi/d$ which is perpendicular to the film surface. The positive integer p denotes the quantization number of the standing spin wave. Frequencies of the PSSW modes can approximately be described by[18]

$$\nu_p = \frac{\gamma}{2\pi}\left[\left(H + \frac{2A}{M_S}q_\parallel^2 + \frac{2A}{M_S}\left(\frac{p\pi}{d}\right)^2\right)\right.$$
$$\left.\times\left(H + \left[\frac{2A}{M_S} + H\left(\frac{4\pi M_S/H}{p\pi/d}\right)^2\right]q_\parallel^2 + \frac{2A}{M_S}\left(\frac{p\pi}{d}\right)^2 + 4\pi M_S\right)\right]^{\frac{1}{2}}, \qquad (3)$$

where A is the exchange stiffness constant. Note that in case of fixed experimental conditions (i.e., H and $q_\parallel$ are constant) and with γ and d being unchanged, the frequency of the DE mode (Eq. (2)) only depends on the saturation magnetization M_S. The frequencies of the PSSW modes (Eq. (3)), however, exhibit an inverse dependence on M_S and, in addition, are a function of A.

Figure 7 shows a representative BLS spectrum recorded at an external magnetic field of 1500 Oe from the Co_2MnSi film annealed at 400 °C. In both the Stokes and anti-Stokes regimes two peaks originating from the magnonic scattering processes are visible. In particular, the peak at around 15 GHz can be attributed to the DE mode, while the peak appearing at approximately 25 GHz results from the excitation of the first perpendicular standing spin wave. The characteristic shape of the BLS spectrum remains unchanged for all the investigated samples. A closer inspection of the spectral peak positions, however, reveals that the frequency of the DE mode decreases by about 1 GHz when T_a is changed from 350 °C to 500 °C while the frequency of the PSSW mode increases by about 1.5 GHz (Fig. 8). Equations (2) and (3) show that the frequency shift of the DE mode and the PSSW mode in Fig. (3) is related to a decrease of the saturation magnetization M_S. However, Eq. (3) indicates that the increase of the PSSW mode frequency might additionally be caused by an increase of the exchange stiffness constant A. From numerical simulations using a theoretical model described in Ref.[19], we estimated that the Co_2MnSi film with the smallest degree of $L2_1$ order exhibits a value of M_S which is about 9 % higher and an exchange stiffness constant which is about by 8 % lower than the corresponding values for the Co_2MnSi sample with the highest amount of the $L2_1$ phase. Unfortunately, as BLS spectra of the investigated Co_2MnSi films only showed a single PSSW mode, the numerical values of A could not be determined.

The saturation magnetization at RT for the Co_2MnSi film annealed at $T_a = 350\,^{\circ}$C could be obtained by means of vibrating sample magnetometry and was found to be $1013\,\mathrm{emu/cm^3}$. This value perfectly agrees with the theoretically predicted value of $M_S = 5.07\,\mu_B$/f.u. (i.e., $1040\,\mathrm{emu/cm^3}$) for the bulk Co_2MnSi with $L2_1$ order[20]. Taking into account the decrease of M_S by 9 % obtained from numerical simulations, the experimentally obtained value of M_S is still in a good agreement with its theoretical counterpart.

The study of spin wave frequencies in dependence of the in-plane sample orientation allows for the determination of anisotropy constants of thin magnetic films. We performed such BLS spectroscopy measurements for the Co_2MnSi films annealed at 350, 375, 400, 450 and 500 °C. All spectra were recorded at a transferred wave vector of $q_{\|} = 1.67\,\mathrm{cm}^{-1}$ and an external magnetic field of $H = 300$ Oe, which was large enough to saturate the sample. The angle α between H and the [110] easy axis direction of the Co_2MnSi films was varied from $0\,^{\circ}$ to $180\,^{\circ}$ in $15\,^{\circ}$ steps by rotating the sample. Due to the cubic symmetry of the crystal structure, the variation of α in the whole range from $0\,^{\circ}$ to $360\,^{\circ}$ was not required.

Figure 9 shows the frequency of the DE mode as function of the external magnetic field H for the Co_2MnSi film annealed at $T_a = 375\,^{\circ}$C. The sample exhibits a clear four-fold magnetic anisotropy which perfectly agrees with the results obtained from the MOKE investigations presented in the previous section. Maxima of the surface spin wave frequencies at $\alpha = n \times 90^{\circ}$, with n being an integer, show that the [110] directions are easy axis directions. Furthermore, from the minima of the DE frequencies at $(n + 1/2) \times 90^{\circ}$ we deduce that the [100] directions are hard axis directions. Fitting the theoretical curve obtained by means of the model described in Ref.[19] to the experimental data, we determined the cubic volume anisotropy constant K_1. The values of K_1 for samples annealed at different temperatures are shown in Fig. 10. The maximal value of $K_1 = -9 \cdot 10^5$ erg/cm^3 was found for $T_a = 375\,^{\circ}$C. The increase of T_a leads to a drop of the volume anisotropy constant by a factor of 10, which might be related to the improvement of the $L2_1$ order. However, other factors such as diffusion of Cr into the Co_2MnSi layer might play a role as well.

V. SUMMARY

The influence of the $L2_1$ ordering degree on the magnetic properties of the Co_2MnSi Heusler compound was investigated. Different amounts of the $L2_1$ phase were realized by

different post-growth annealing temperatures ranging from 350 °C to 500 °C. Summarizing the results of the magnetic characterization using MOKE and BLS we reveal that an increased $L2_1$ ordering degree of the Co_2MnSi films is correlated with a reduction of the saturation magnetization of about 9 %, an increase of the exchange stiffness constant of 8 % and a drop of the cubic anisotropy constant K_1 by a factor of 10.

VI. ACKNOWLEDGMENT

The project was financially supported by the Research Unit 559 *"New materials with high spin polarization"* funded by the Deutsche Forschungsgemeinschaft and by the NEDO International Joint Research Grant Programm 2004/T093. We would like to thank M. Jourdan, H. Schneider and H. Schultheiß for fruitful discussions and technical advice.

[1] I. Galanakis, P. H. Dederichs, and N. Papanikolaou, Phys. Rev. B **66**, 174429 (2002).

[2] S. Picozzi, A. Continenza, and A. J. Freeman, Phys. Rev. B **66**, 094421 (2002).

[3] P. J. Brown, K.-U. Neumann, P. J. Webster, and K. R. A. Ziebeck, J. Phys.: Condens. Matter **12**, 1827 (2000).

[4] S. Fuji, S. Sugimura, S. Ishida, and S. Asano, J. Phys.: Condens. Matter **2**, 8583 (1990).

[5] J. Schmalhorst, S. Kämmerer, M. Sacher, G. Reiss, and A. Hütten, Phys. Rev. B **70**, 024426 (2004).

[6] Y. Sakuraba, J. Nakata, M. Oogane, H. Kubota, Y. Ando, A. Sakuma, and T. Miyazaki, Jpn. J. Appl. Phys. **44**, L1100 (2005).

[7] Y. Sakuraba, M. Hattori, M. Oogane, Y. Ando, H. Kato, A. Sakuma, T. Miyazaki, and H. Kubota, Appl. Phys. Lett. **88**, 192508 (2006).

[8] Y. Sakuraba, T. Miyakoshi, M. Oogane, Y. Ando, A. Sakuma, T. Miyazaki, and H. Kubota, Appl. Phys. Lett. **89**, 052508 (2006).

[9] W. H. Wang, M. Przybylski, W. Kuch, L. I. Chelaru, J. Wang, Y. F. Lu, J. Barthel, H. L. Meyerheim, and J. Kirschner, Phys. Rev. B **71**, 144416 (2005).

[10] S. Picozzi, A. Continenza, and A. J. Freeman, Phys. Rev. B **69**, 094423 (2004).

[11] S. Okamoto, O. Kitakami, and Y. Shimada, J. Magn. Magn. Mater. **208**, 102 (2000).

[12] K. Postava, H. Jaffres, A. Schul, F. N. V. Dau, M. Goiran, and A. R. Fert, J. Magn. Magn. Mater. **172**, 199 (1997).

[13] R. Mattheis and G. Quednau, J. Magn. Magn. Mater. **205**, 143 (1999).

[14] T. Mewes, H. Nembach, M. Rickart, and B. Hillebrands, J. Appl. Phys. **95**, 5324 (2004).

[15] J. Hamrle, S. Blomeier, O. Gaier, B. Hillebrands, K. Postava, H. Schneider, G. Jakob, and C. Felser, J. Phys. D: Appl. Phys. **40**, 1563 (2007).

[16] J. Hamrle, S. Blomeier, O. Gaier, B. Hillebrands, R. Schäfer, and M. Jourdan, J. Appl. Phys. **100**, 103904 (2006).

[17] B. Hillebrands, Rev. Scien. Instr. **70**, 1589 (1999).

[18] S. O. Demokritov, B. Hillebrands, and A. N. Slavin, Physics Reports **348**, 441 (2001).

[19] B. Hillebrands, Phys. Rev. B **41**, 530 (1990).

[20] P. J. Webster, J. Phys. Chem. Solids **32**, 1221 (1971).

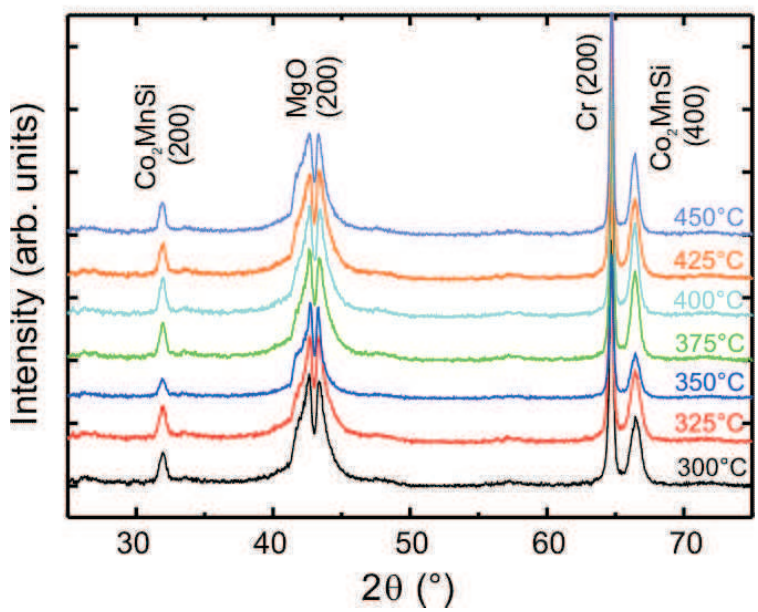

FIG. 1: (Color online) θ-2θ-scans of MgO/Cr(40 nm)/Co_2MnSi(30 nm)/Al(1.3 nm) films annealed at temperatures ranging from 350 °C to 500 °C.

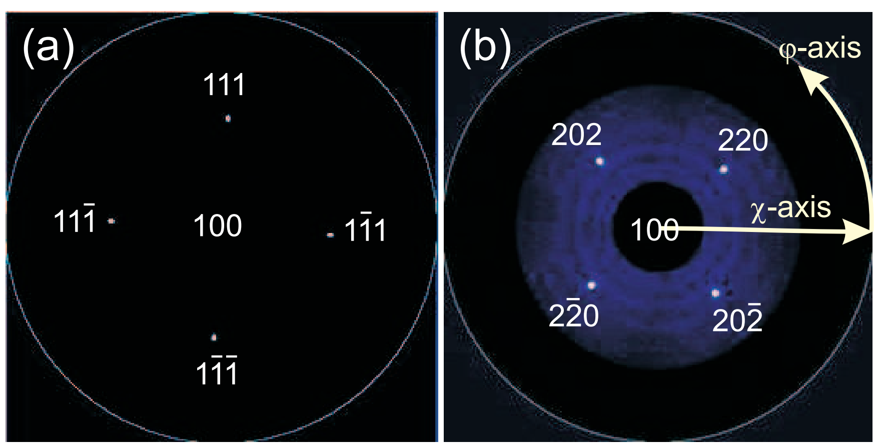

FIG. 2: (Color online) X-ray pole figures for (a) (111)-planes and (b) (220)-planes of the Co_2MnSi film annealed at $T_a = 450$ °C.

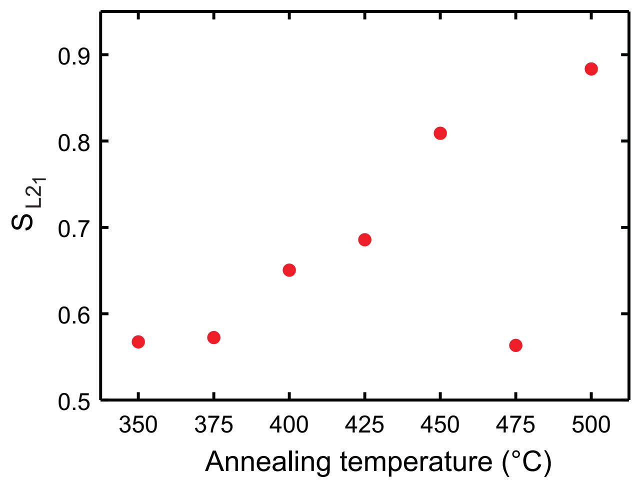

FIG. 3: (Color online) Long-range order parameter S_{L2_1} for Co_2MnSi films annealed at different temperatures after the deposition. The experimental error of S_{L2_1} values corresponds to the size of the data points. Note that $S_{L2_1} = 1$ for a perfect order on the Mn and Si sites.

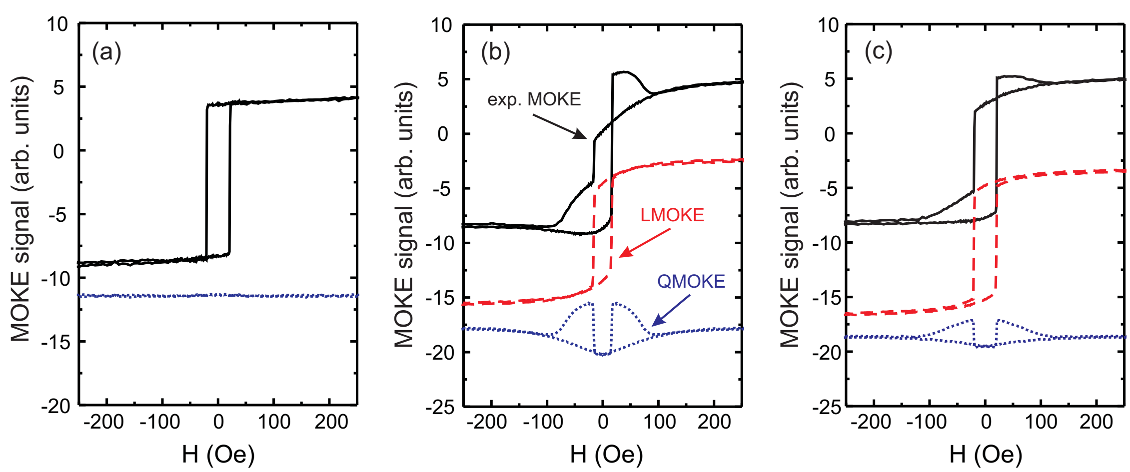

FIG. 4: (Color online) Room temperature hysteresis loops measured by magneto-optical Kerr effect (MOKE) magnetometry at sample orientations α of (a) 0°, (b) 42° and (c) 45° for the Co_2MnSi film annealed at 425 °C (full line). The symmetrization and antisymmetrization of these loops provide the LMOKE (dashed line) and QMOKE (dotted line) contributions.

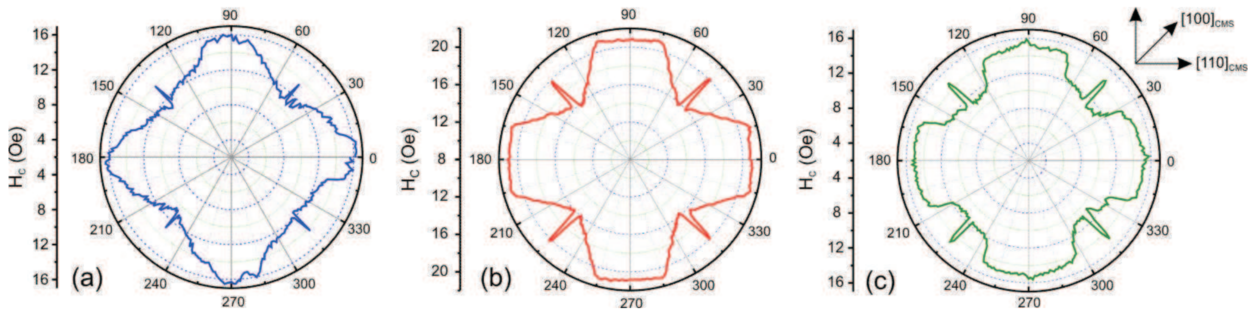

FIG. 5: (Color online) Angular dependence of the room temperature coercive field H_C for Co_2MnSi films annealed at (a) 350 °C, (b) 425 °C and (c) 500 °C. α denotes the angle between the external magnetic field H and the [110] crystallographic direction of the Co_2MnSi films. Note the different scale for the annealing temperature of 425 °C.

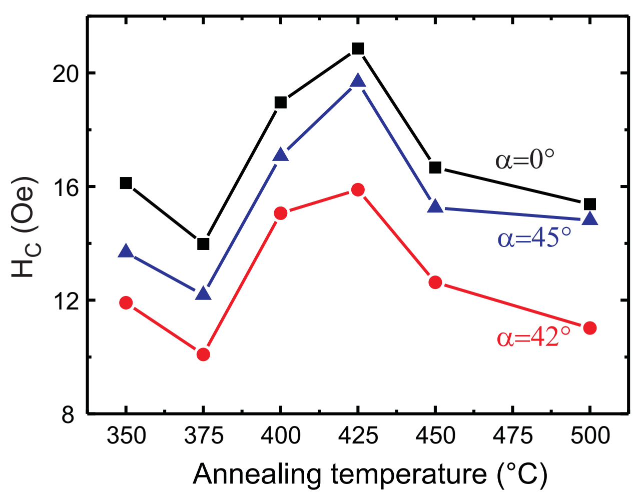

FIG. 6: (Color online) Variation of the room temperature coercive field H_C with the annealing temperature for sample orientations $\alpha = 0°$, 42° and 45°.

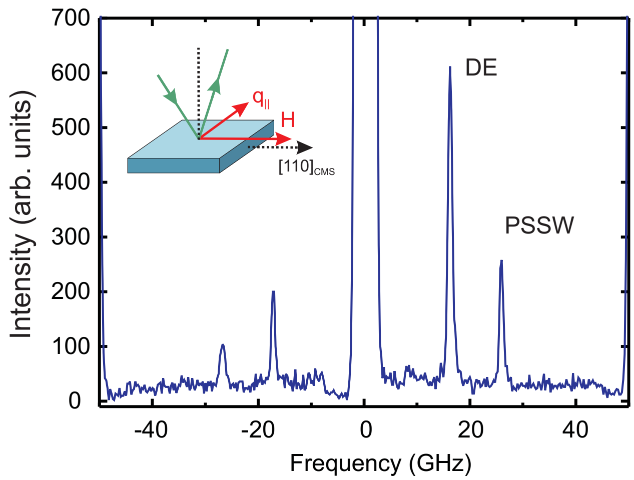

FIG. 7: (Color online) BLS spectrum of the Co_2MnSi film annealed at 400 °C, measured at an applied magnetic field of $H = 1500\,\mathrm{Oe}$ and a transferred wave vector of $q_{\|} = 1.67\,\mathrm{cm}^{-1}$. H was aligned parallel to the [110] direction of the Co_2MnSi sample, as sketched in the inset.

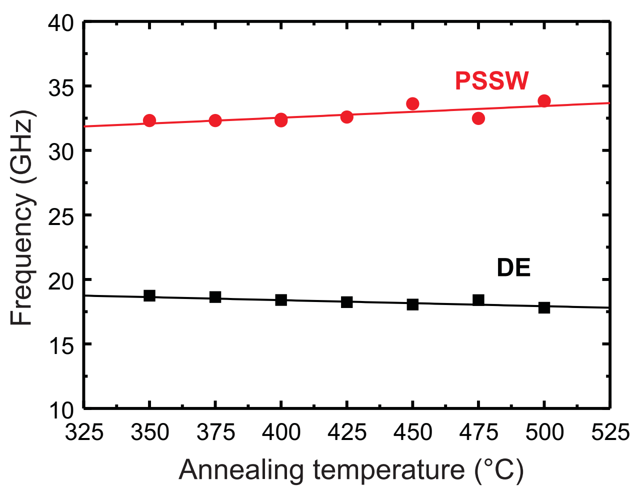

FIG. 8: (Color online) Dependence of room temperature spin wave frequencies on the annealing temperature of the Co_2MnSi films. The frequencies were determined from BLS spectra measured at an applied magnetic field of $H = 2000\,\mathrm{Oe}$ and a transferred wave vector of $q_{\|} = 1.67\,\mathrm{cm}^{-1}$.

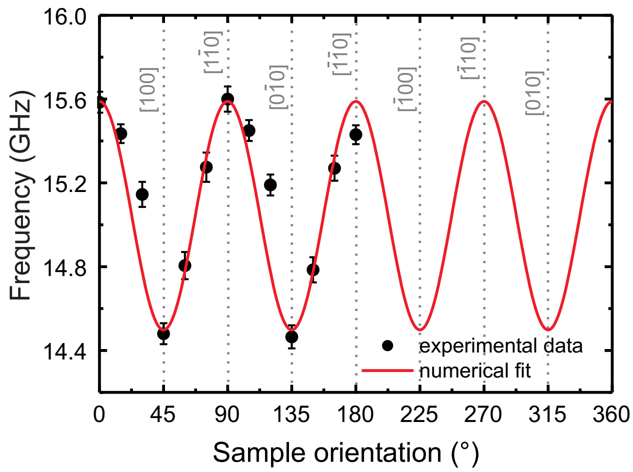

FIG. 9: (Color online) Frequency of the DE mode as function of the angle between the external magnetic field $H = 300\,\mathrm{Oe}$ and the [110] easy axis direction for the Co_2MnSi film annealed at $375\,^\circ\mathrm{C}$.

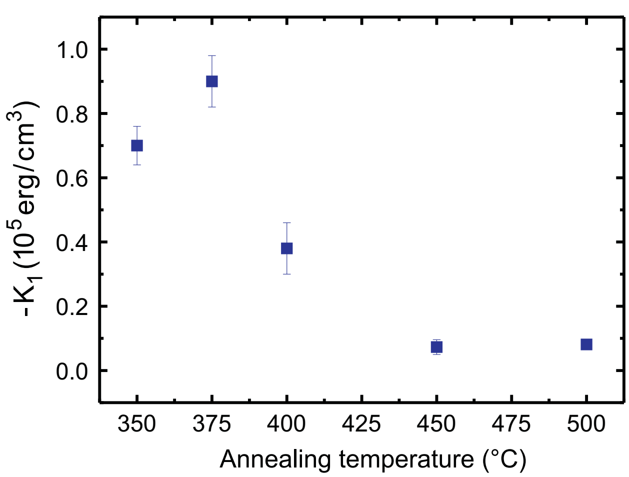

FIG. 10: (Color online) Room temperature volume anisotropy constant K_1 for Co_2MnSi films annealed at different temperatures T_a.